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Enhancement of the light extraction of GaN-based green light emitting diodes via nanohybrid structures

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A B S T R A C T

Improvement in the light extraction efficiency (LEE) of GaN-based green light emitting diodes (LEDs) with ZnO nanostructures synthesized by a hydrothermal method is reported. Formation of ZnO nanorods, hemispheres, and cones was controlled by varying the pH of the aqueous synthesis solution. The shape of the ZnO nanostructures integrated onto the LEDs shows a strong relationship with the LEE characteristics of GaN-based green LEDs. The electroluminescence (EL) intensity of LEDs covered by ZnO nanostructures increased compared to conventional LEDs. In terms of LEE, LEDs with surface-textured ZnO hemispheres showed the highest EL intensity, which can be attributed to an increase in the effective critical angle, the escape cone, and multiple scattering. Finite difference time domain (FDTD) simulation was conducted to theoretically confirm the experimental results.

1. Introduction

Remarkable technological improvements in GaN-based light emitting diodes (LEDs) in the past two decades have inspired many research groups to focus on the replacement of conventional lighting sources with high efficiency LEDs. GaN-based LEDs are promising candidates for lighting sources and are widely used in various optoelectronic applications, such as full color displays, automobiles, and backlight units for liquid crystal displays, due to their high efficiency, long lifetime, and low power consumption [1]. In order to produce high-performance LEDs with the long-term reliability of a conventional bulb, it is critical to increase the quantum efficiency of GaN LEDs. In terms of the development of full color displays, increasing the performance LEDs of all three primary colors—red, green, and blue—is essential. Although InGaN-blue and AlGAInP-red LEDs with relatively high quantum efficiency have been developed, the quantum efficiency of green LEDs is still less than what is available for other visible-range LEDs [2,3]. The relatively low quantum efficiency of green LEDs can be attributed to the difficulty of inducing the growth of high-quality InGaN active layers with a high indium composition without degrading the crystal quality. By increasing the indium composition, the composition pulling and high piezoelectric field effects are increased in InGaN/GaN multiple quantum wells (MQWs), resulting in deterioration of the material properties and LED performance [3].

Promising results in increasing the internal quantum efficiency (IQE) and light extraction efficiency (LEE) of GaN-based LEDs in order to improve the light output power of LEDs have been reported. The use of techniques such as surface texturing and the incorporation of photonic crystals, patterned sapphire substrates (PSS), and V-pit embedded structures have been suggested, some of which are currently used in mass production lines [4–8]. Recently, efforts to produce nanohybrid structures through the integration of nanomaterials with GaN LEDs structures have been conducted. To form nanostructures on top of LED structures, candidate materials such as ZnO, SiO_2/polystyrene, indium-tin-oxide (ITO), MgZnO, and TiO_2 have been suggested [9–12]. Among these materials, ZnO nanostructures have been extensively investigated for increasing the quantum efficiency of GaN-based LEDs. Many research groups have reported that the morphological control of ZnO nanorods and nanocones on LED epitaxial structures can improve the LEE in GaN LEDs [13,14].

We investigated the effects of changes in the surface morphology of ZnO nanostructures formed on p-GaN surfaces on the extraction efficiency of a green-emission LED. The changes in the morphology of ZnO nanostructures grown by using a hydrothermal method have strong influence on the light extraction of InGaN-based green LEDs. Theoretical investigations were

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conducted using Lumerical FDTD simulations in order to support the experimental results.

2. Experimental details

Epitaxially-grown LED structures were produced using a conventional metal organic chemical vapor deposition (MOCVD) system. Schematic pictures of the ZnO nanostructures formed on epitaxial GaN LED structures are shown in Fig. 1. ZnO nanostructures were synthesized on the green epitaxial LED templates via a hydrothermal method due to the low cost and simplicity. In order to obtain nanostructure arrays, a ZnO seed layer was formed on top of the LED templates before hydrothermal synthesis. The substrate was then dipped and the reaction was conducted in zinc nitrate hexahydrate (ZNH, $[Zn(NO_3)_2 \cdot 6H_2O]$) and hexamethylenetetramine (HMT, $[C_6H_{12}N_4]$) dissolved in deionized (DI) water at 95 °C for 6 h as described previously [15]. To control the morphology of ZnO, pH values of the aqueous solution were varied from 7 to 12 by adjusting using ammonium hydroxide. After synthesis of the morphology-controlled ZnO nanostructures on the LED templates, the samples were rinsed with DI water and dried by blowing with nitrogen (N_2) gas at room temperature. To measure the electroluminescence (EL) of LED structures with different ZnO nanostructures, the indium pad was transferred to the p-and n-regions of the GaN-based LEDs. The morphology and microstructures of the ZnO nanostructures on the LED templates were characterized by scanning electrode microscopy (SEM; Hitachi S-4800). Electrical characterizations were conducted using electroluminescence (EL; LIV system/LEOS OPI-150-Optel-precision) to investigate the effects of the surface morphology of ZnO nanostructures on the LEE characteristics of green LEDs.

3. Results and discussion

Fig. 2 shows 15°-tilted SEM images of various ZnO nanostructures formed on top surface of LED structures. Surface morphology was controlled by varying the pH, and pH values of 7, 10, and 12 were obtained by varying the amount of ammonium hydroxide in the aqueous solutions. All samples show vertically-aligned ZnO nanostructures on the epitaxial GaN LED templates and high nanostructure density. Fig. 2(a) shows the typical hexagonal ZnO nanorod structures synthesized in a pH 7 solution. By increasing the pH to 10, the morphology of the ZnO nanostructures changed from nanorods to hemispheres covered with very small ZnO nanorods, as shown in Fig. 2(b). The magnified image of the hemisphere structure clearly shows bundles of very small nanowires covering the surface. Because of this covering of tiny nanowires, the surface area is expected to be greater than that of the hexagonal nanorods formed at pH 7. The surface morphology of the ZnO structures changed from hemispheres to nanocones with an increase in pH to 12. Variation of the pH of the aqueous solution had very strong effects on the surface morphology of the ZnO nanostructures [16,17]. However, it is unknown what the dominant mechanism responsible for this change in the surface morphology of ZnO nanostructures is, and more study is required to elucidate this process. Fig. 2(d) shows the p-GaN surface of conventional GaN-based LEDs without hydrothermal synthesis.

EL measurements were conducted to investigate the ZnO nanostructures formed on the p-GaN surface of GaN LED structures. The EL emission peak wavelengths of the GaN green LEDs were located at 520–530 nm as shown in Fig. 3. Compared to conventional LEDs without nanostructures, the EL intensities obtained from the LED structures increased by 4.97, 5.87, and 6.70 times for

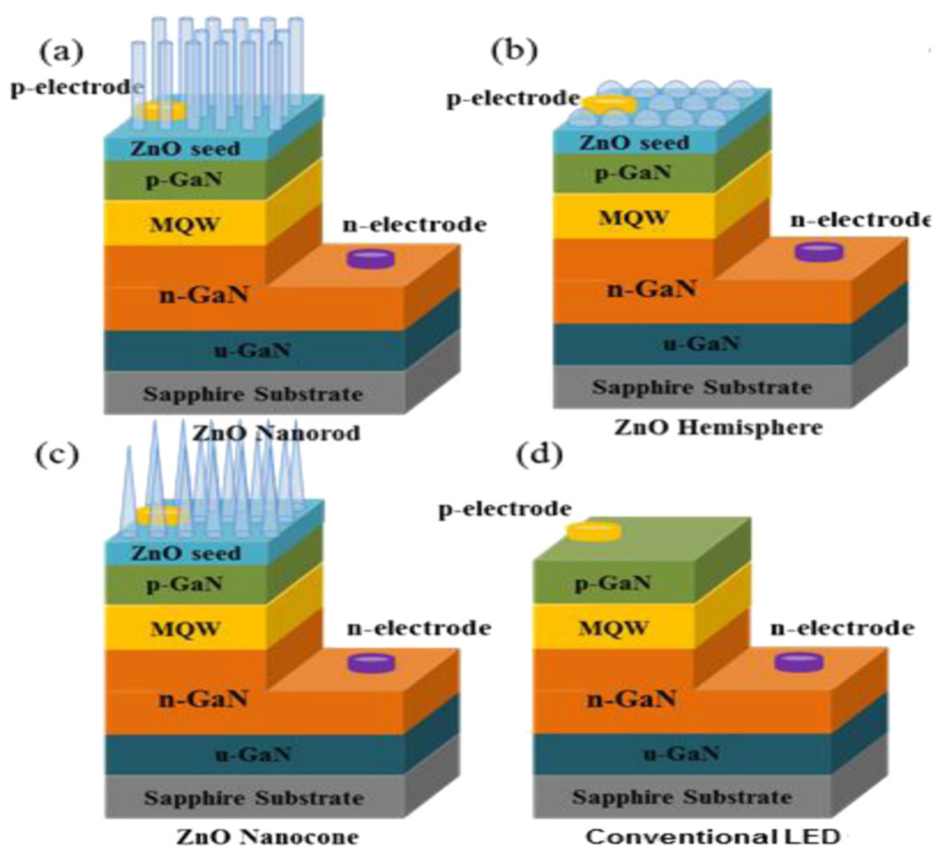

Fig. 1. Schematic configuration of LED epitaxial structures with (a) nanorods, (b) hemispheres, (c) nanocones, and (d) without nanostructures (conventional LED).